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Differential-Die Away Instrument: Report on Initial Simulations of Spent Fuel Experiment

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Abstract

New Monte Carlo simulations of the differential die-away (DDA) instrument response to the assay of spent and fresh fuel helped to redefine the signal-to-background ratio and the effects of source neutron tailoring on the system performance. Previously, burst neutrons from the neutron generator together with all neutrons from a fission chain started by a fast fission of ^{238}U were considered to contribute to active background counts. However, through additional simulations, the magnitude of the ^{238}U first fission contribution was found to not affect the DDA performance in reconstructing $^{239}Pu_{eff}$. As a result, the newly adopted DDA active background definition considers now any neutrons within a branch of the fission chain that does not include at least one fission event induced by a thermal neutron, before being detected, to be the active background. The active background, consisting thus of neutrons from a fission chain or its individual branches composed entirely of sequence of fast fissions on any fissile or fissionable nuclei, is not expected to change significantly with different fuel assemblies. Additionally, while source tailoring materials surrounding the neutron generator were found to influence and possibly improve the instrument performance, the effect was not substantial.

Introduction

The National Nuclear Security Administration (NNSA) Office of Nonproliferation and Verification Research and Development provided funding to advance the capability of the DDA instrument. The NNSA Office of Nonproliferation and International Security initiated the Next Generation Safeguards Initiative (NGSI) Spent Fuel Project in order to develop non-destructive assay techniques specifically for assaying spent fuel assemblies [1]. The technical goals of the project have evolved to include the following: (1) determine total Pu mass, (2) estimate basic fuel assembly parameters such as initial enrichment (IE), burn-up (BU) and cooling time (CT), (3) quantify the degree to which the fuel assembly is intact (partial defect test) and (4) quantify the heat content of the assembly [2]. In pursuit of these goals, this research focuses on one of these NDA techniques, the differential die-away (DDA) that will eventually be integrated with other measurement techniques to form a measurement system.

Using a short pulse of high-energy neutrons from an external deuterium-tritium (DT) neutron generator, the DDA technique actively interrogates a fuel assembly leading to the release of prompt neutrons primarily from the fission of fissile material in the spent fuel assembly (SFA). As the system is subcritical, the induced neutron generations caused by the neutron generator and the multiplying material die away on the order of hundreds of microseconds. The measured DDA signal reveals properties of the fuel assembly, primarily multiplication (M) [3], and is implicitly a function of the IE, BU, and CT [4].

In early FY2014, Los Alamos National Laboratory's Nuclear Engineering and Nonproliferation Safeguards Science & Technology Group (NEN-1) performed initial simulations of the planned DDA instrument experimental measurements. A simpler DDA system than originally modeled by Blanc et al [5] was simulated using Monte Carlo N-Particle eXtended (MCNPX) v274 [6] to study the effects of source neutron tailoring on instrument performance. Originally, the DDA active background, present only during the active interrogation and virtually independent of the spent fuel assembly (SFA), was defined as neutrons reaching the detectors directly from the neutron generator (burst neutrons) and any neutrons that were from a fission chain started by a fast fission of uranium-238 (^{238}U). However with new simulations of both fresh fuel and spent fuel, a new understanding of the DDA background was surmised. Instead, in addition to the burst neutrons, only neutrons from a branch of a fission chain that does not include at least one fission event induced by a thermal neutron to be part of the active background. This new definition stems from the principle of the assay that is primarily sensitive to fissile content by means of fission induced by thermalized neutrons. Also, from the perspective of the fast neutrons in the interrogation process, the effect of the isotopic composition of individual SFAs (mainly ^{238}U) is not important.

This new understanding also led to new parameters for evaluating the DDA performance dependence on the quality and quantity of the tailoring material between the neutron generator and the SFA. The three parameters calculated in simulations dedicated to the source neutron tailoring study were (1) the fraction of detected fission neutrons, defined as the number of detected fission neutrons per the number of detected burst neutrons, (2) the detected fission efficiency, defined as the number of detected fission neutrons per the total number of neutrons emitted from the neutron generator, and (3) the ^{238}U first fission fraction, defined as the number of fission chains started by fission of ^{238}U per the number of all fission chains.

Spent Fuel: Simulations of the Impact of Source Neutron Tailoring

In the original DDA designs [5], large quantities of source tailoring materials were used in the DDA instrument to moderate the energy of neutrons from the neutron generator below the 1 MeV energy threshold of ^{238}U fission and possibly increase the interrogating neutron flux through (n,2n) reactions such as in tungsten (W) (Fig. 1). However, our new understanding of DDA active background indicates that the first fission of ^{238}U nuclei in the fuel assembly does not negatively impact the DDA performance, so long as the neutrons emitted from ^{238}U lead to additional thermal induced fissions in the fissile nuclides (^{235}U, ^{239}Pu, ^{241}Pu) of the fuel.

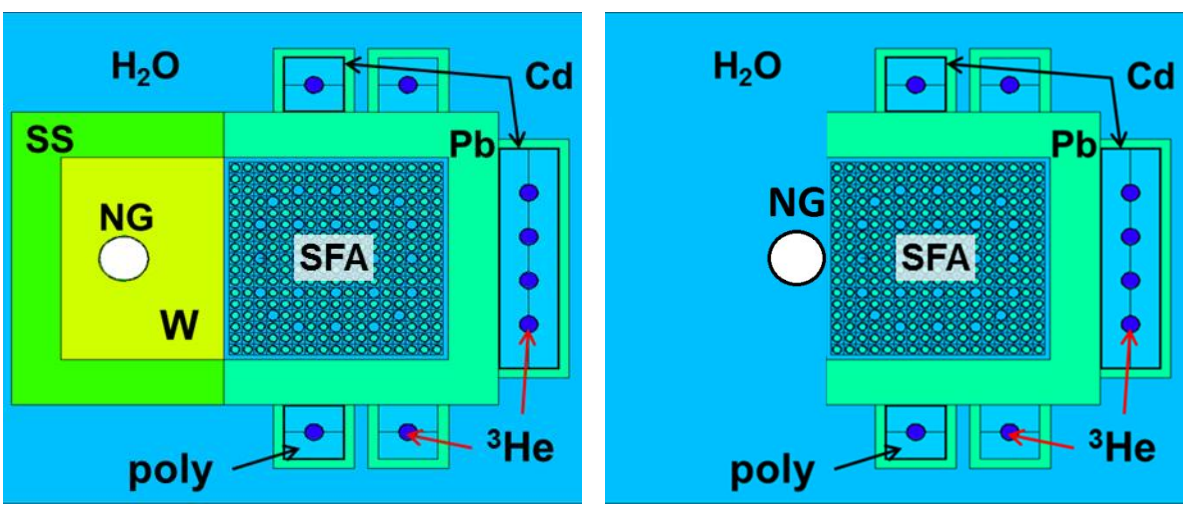

Figure 1. *(Left)* Blanc et al DDA design [5] and *(right)* simplified "bare" source tailoring test design

As an ultimate test of this hypothesis, we simulated a stripped down ("bare") version of the original DDA setup with spent fuel in water, without any source tailoring material between the fuel and neutron generator, and the neutron generator positioned as close to the fuel assembly as possible. Eight helium-3 (^{3}He) neutron detectors encapsulated in high-density polyethylene (HDPE) blocks wrapped with cadmium (Cd) were simulated. The purpose of these simulations was to keep the neutron spectrum entering the SFA as hard as practically achievable, thus increasing the fraction of ^{238}U first fission. As a consequence, the burst neutron contribution to the active signal was significantly increased by approximately a factor of 3 on the back detectors in the time domain of 100-200 μs (an increase from 6.0% contribution in the original Blanc et al design to 19% in the "bare" version). The contribution of detected neutrons from fission chains started by fissions on ^{238}U to the DDA signal in the same time domain increased from 5.8% in the Blanc et al design to 34.2% in the "bare" version in the particular case a SFA with 4% IE, 45 GWd/tU BU and 5 y of CT from Spent Fuel Library-2a (SFL-2a) [7]. However, even with such a profound increase to the contribution of first fission on ^{238}U, there was no noticeable deterioration of the DDA performance measured by its ability to reconstruct the plutonium-239 effective ($^{239}Pu_{eff}$) (Fig. 2) as compared to results for SFL-1, SFL-2 and SFL-2e where the original Blanc et al DDA design with a massive tungsten block as tailoring material was simulated. The $^{239}Pu_{eff}$ is defined [8] as the corresponding mass of ^{239}Pu that would give the same signal response as that obtained from all fissile isotopes (^{235}U, ^{239}Pu, and ^{241}Pu) in the fuel assembly and thus is an effective measure of the total fissile content in the SFA.

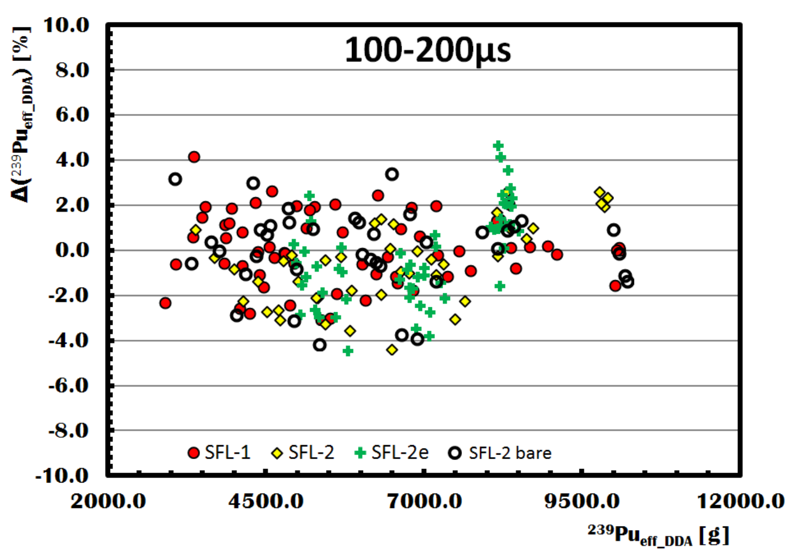

Figure 2. The results of the percent difference of $^{239}Pu_{eff}$ reconstruction for the "bare" DDA design compared to the other SFL simulations using the original Blanc et al design with a tungsten block as the source neutron tailoring material.

Confirming our new hypothesis, these results indicate that the first fission on ^{238}U does not represent a serious issue that would decisively hinder the performance of the DDA instrument. However, as we present below, source tailoring material or lack thereof still may change the overall performance of the instrument. These effects were investigated in additional MCNPX simulations using fresh fuel.

Fresh Fuel: Simulations of the Impact of Source Neutron Tailoring on DDA Signal

Multiple source tailoring material types and thicknesses were simulated using MCNPX v274 for a 15x15 fresh fuel assembly (FFA) with 1.96% ^{235}U enrichment submerged in water and surrounded by nine ^{3}He neutron detectors encapsulated in HDPE cylinders wrapped with Cd, with a DT neutron generator positioned in air (Fig. 3). The source tailoring materials simulated were iron (Fe), water (H_2O), lead (Pb), tungsten (W), and air in 2 cm, 4 cm, and 8 cm thicknesses with the balance filled with air because the neutron generator position remained constant for all runs. All simulations used a 20 μs long interrogating pulse from the NG, to compare with experiments to be carried out in the near future. (For DDA experiments, the DT neutron generator operates at 2500 Hz with a 5% duty cycle, resulting in a 20 μs interrogation pulse.)

The total detector signal was tallied along with a subdivision of this tally for individual detectors. The detected first fission contribution from individual isotopes (only uranium isotopes for the case of fresh fuel), and the detected neutrons directly from the DT neutron generator were tallied to indicate their origin. In the case of the fissile isotopes, a capability added to the MCNPX code explicitly for the NGSI project and not available in standard public release versions, called "first fission," was used. In these simulations all neutrons start in the neutron generator. If the neutron, as it advances through various materials of the DDA instrument in the simulation should cause a fission, the identifier on the emitted neutron(s) is changed from originating in the neutron generator to originating in the isotope that underwent fission. From that time onward, the neutron maintains the label it received on its first fission.

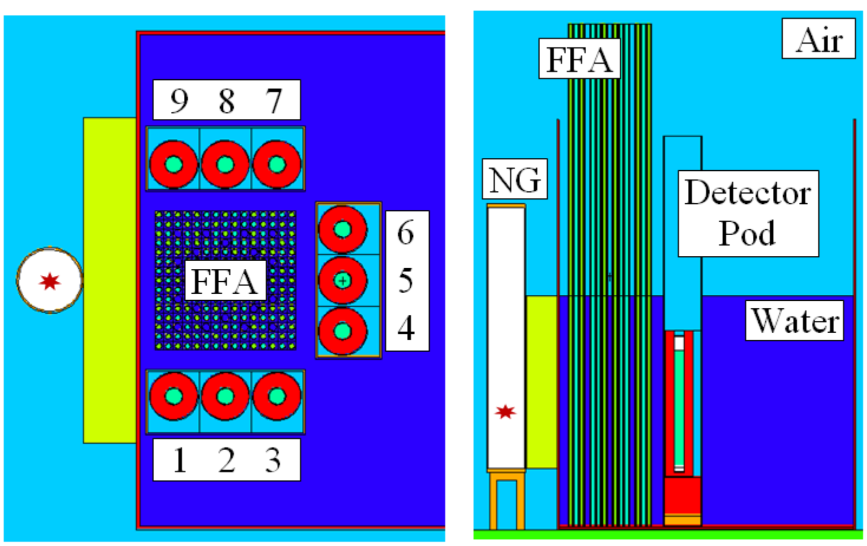

Figure 3. Cross sectional views of the MCNPX DDA instrument simulations with 8 cm of source tailoring material (green block) between the neutron generator (NG) and the fresh fuel assembly (FFA)

The three quantities calculated for all source tailoring arrangements were (1) the fission-to-burst ratio, or the number of detected neutrons from the fission of ^{235}U and ^{238}U per the number of detected burst neutrons, (2) the fission-to-source ratio (detected fission efficiency), or the number of total fission neutrons per the total source neutrons emitted from the neutron generator, and (3) the ^{238}U first fission fraction, or the detected neutrons which came from the fission of ^{238}U to total (^{235}U and ^{238}U) fission ratio was calculated to determine the impact of the source tailoring material on the response of the DDA instrument. Primarily for historical context, the ^{238}U first fission fraction was evaluated even though results from the SFA DDA simulations show that it does not affect the ability of the DDA instrument to reconstruct $^{239}Pu_{eff}$. Thus, the ^{238}U first fission fraction may be deemed an irrelevant quantity for the evaluation of the DDA instrument performance in the future.

The fission-to-burst ratio, which describes the DDA signal neutrons which originated from fission on ^{235}U and ^{238}U per the detected neutron generator burst neutrons, was calculated for all source tailoring cases for each detector around the FFA (Fig. 4). However, recent DDA findings by Martinik et al [9] show that the back detectors are instrumental for the proper overall characterization of the assayed fuel assembly. In the following section, an evaluation of the source tailoring will focus particularly on the DDA signal from the back detectors. In the future, further assessment of the front detector signals relative to the back detector signals will be performed to potentially determine additional information about specific regions, including the presence of partial defects, within the fuel assembly. As expected, the detectors located closest to the neutron generator recorded the smallest fission-to-burst ratio. The back detector ratios varied between 1.7 and 4.8 for air and tungsten, respectively. The effect on the fission-to-burst ratio from iron and lead was nearly the same for each material thickness. Tungsten produced the largest effect on the back center detector (Detector 5), but did not yield a substantial improvement over the other high density (Pb, Fe) source tailoring materials. The improvement in signal-to-background between the worst and best source tailoring arrangement was approximately a factor of 2.5.

The fission-to-source ratio, or the detected fission efficiency, is a ratio of all detected fission neutrons over all neutrons emitted by the neutron generator whether they cause fission or not and whether they are detected or not. The upper and lower detectors recorded a larger fission-to-source ratio, implying that a greater number of fissions were induced in the volume of the fuel assembly closer to the neutron generator (Fig. 5). Then as neutrons from first induced fission propagate through the fuel assembly, the overall neutron flux weakens, even though additional fissions are induced, due to the subcritical fuel assembly. Neutrons from induced fissions in the back region of the FFA are predominantly those that get detected in the back detectors. Overall, for the back detectors the detected fission efficiency varied between $1.1\cdot10^{-4}$ and $2.2\cdot10^{-4}$ for Fe and air, respectively. For the back detectors (i.e. Detectors 4-6), less source tailoring increased the detected fission efficiency, while the opposite was true for the very front detectors (Detector 1 and 9), with the exception of water. However, neither source tailoring material nor its thickness caused a substantial change to the detected fission efficiency.

The ^{238}U first fission fraction in the 0-400 μs time domain varied between 10% and 40% showing that the choice of source tailoring material does significantly influence the number of first fissions occurring on ^{238}U by affecting the interrogating neutron spectrum (Fig. 6). The trend of less and lower density source tailoring material corresponding to a larger ^{238}U first fission contribution was observed for all detectors. However, as stated above, the magnitude of the ^{238}U first fission contribution does not affect the overall performance of the DDA instrument and is therefore not a critical design factor.

Due to the results on the unimportance of ^{238}U first fission fraction, the effects from source tailoring material can be described solely by its influence on the fission-to-burst ratio and the fission-to-source (detected fission efficiency) ratio. Therefore we propose a new definition of a figure of merit (FOM) that is constructed as a product of the two ratios (Fig. 7) and which describes the influence of the tailoring material on the signal-to-background (S/B) ratio in case of the DDA instrument. Based on these values, the thickness of Fe and water in the 2 cm to 8 cm range does not influence the performance of the DDA system. However, the thicker the Pb or W (within the limited thicknesses used in this study), particularly for the back detectors, the higher the FOM of the DDA signal. Overall, for all the detectors, the potential improvement in S/B between the worst and best source tailoring arrangements was less than a factor of 2. This indicates that while the choice of the source tailoring material and its thickness may impact the performance of the DDA instrument, it does not seem to be a critical design feature determining success or failure of the instrument as whole.

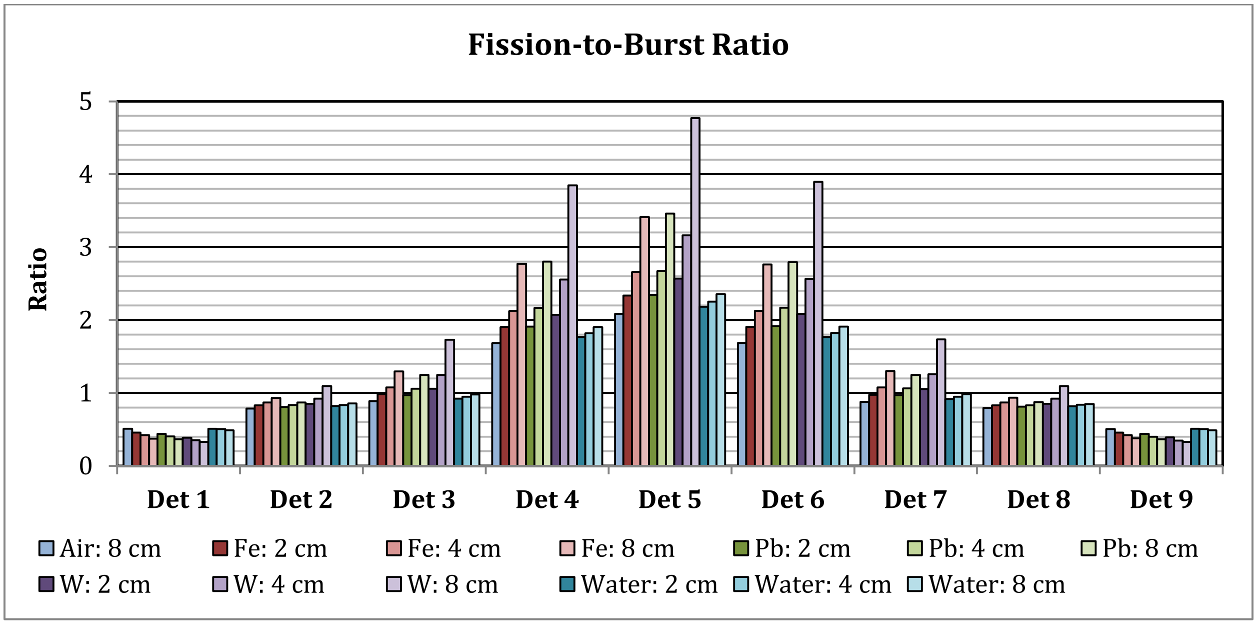

Figure 4. Using the first fission tally results, the detected fission-to-burst DDA signal ratio was calculated for all detectors and all source tailoring schemes in the 0-400 μs time domain

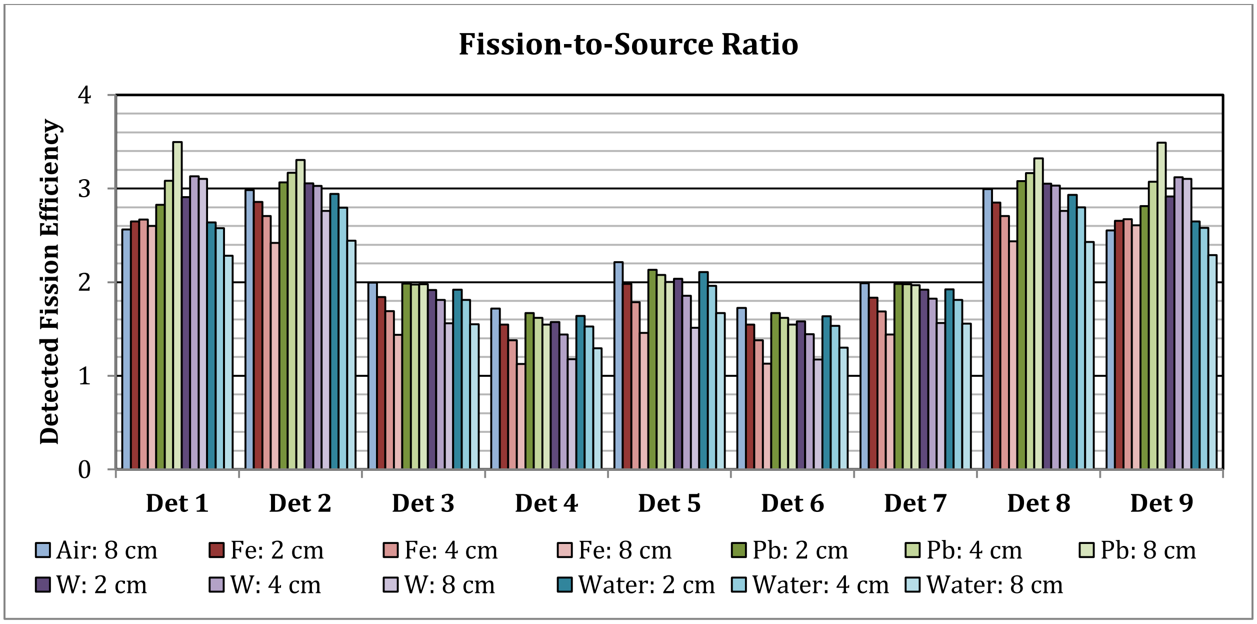

Figure 5. Using the first fission tally results, the fission neutron detection efficiency was calculated for all detectors and all source tailoring schemes in the 0-400 μs time domain

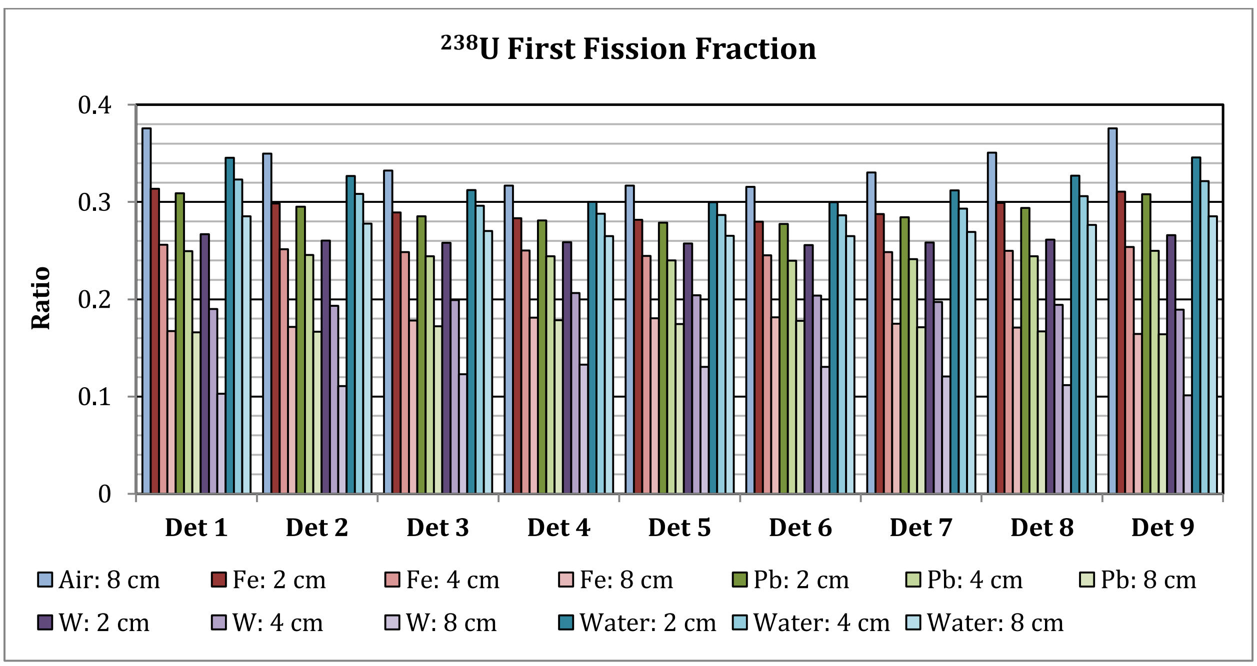

Figure 6. Using the first fission tally results, the ^{238}U first fission fraction was calculated for all detectors and all source tailoring schemes in the 0-400 μs time domain

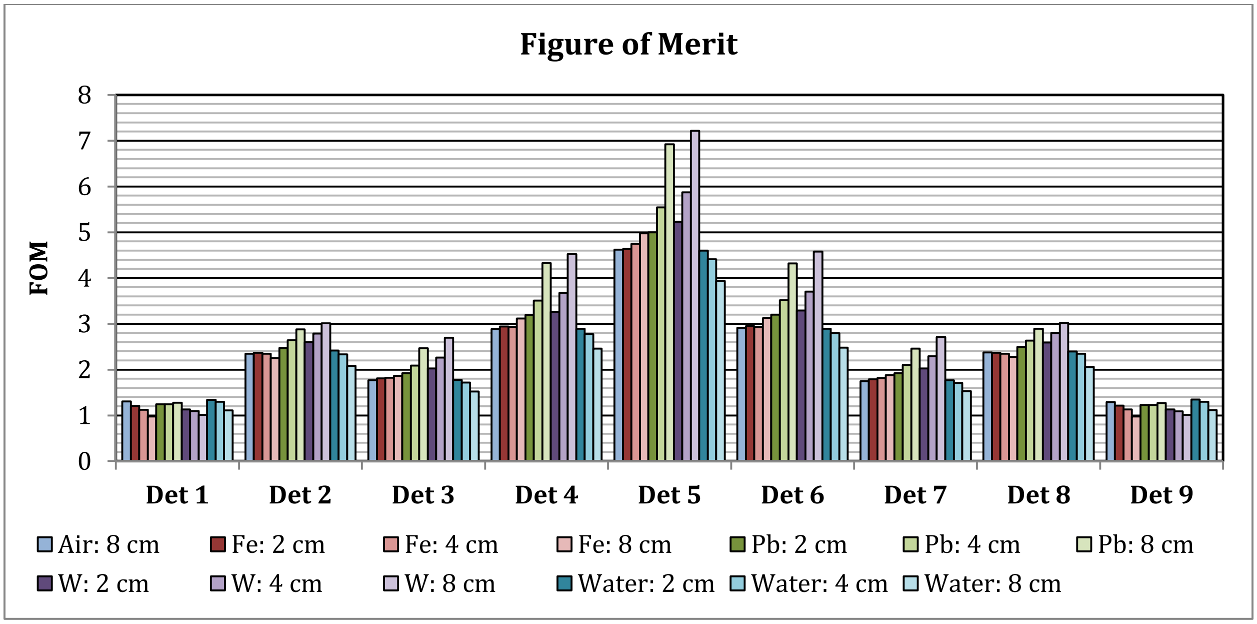

Figure 7. A figure of merit to evaluate the source tailoring effects on the DDA signal was constructed as a product of the fission-to-burst and fission-to-source ratios

Conclusions

Recent Monte Carlo simulations of the DDA system response to active interrogation of fresh and spent fuel led to a new understanding of the active background and the effects of source tailoring on the overall system performance. Our results suggest that the presence of source neutron tailoring material does not substantially influence the DDA signal, as represented by the fission-to-burst and fission-to-source ratios. Based on the calculation of newly proposed FOM, we expect that the overall S/B ratio can be improved by less than a factor of 2 when comparing the best (8 cm thick W) and the worst (air) tailoring material configurations.

As in previous studies, we confirm that the choice of the source tailoring material has significant influence on the ^{238}U first fission contribution. Originally, neutrons from the first fission of ^{238}U were considered as contributors to the active background and thus led to design choices intended to minimize ^{238}U first fast fission. However, in the new DDA background definition, any neutrons which have not undergone at least one fission induced by a thermal neutron on a branch of its fission chain before being detected are considered as contributing to the active background. With dedicated simulations of a “bare” DDA design, we demonstrated that the magnitude of the ^{238}U first fission contribution does not affect the DDA performance gauged by its ability to reconstruct $^{239}Pu_{eff}$ which reflects the fissile content in a given SFA. Therefore, even though tailoring the energy of the source neutrons may help improve the overall performance of the DDA instrument, it should not be considered a critical design factor.

Acknowledgements

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References

[1] M. Humphrey, S. Tobin and K. Veal, "The Next Generation Safeguards Initiative's Spent Fuel Nondestructive Assay Project," *Journal of Nuclear Materials Management*, vol. 40, no. 3, 2012.

[2] S. Tobin, H. Menlove, M. Swinhoe and M. Schear, "Next Generation Safeguards Initiative Research to Determine the Pu Mass in Spent Fuel Assemblies: Purpose, Approach, Constraints, Implementation, and Calibration," *Nuclear Instruments and Methods in Physics Research A*, vol. 652, no. 1, pp. 73-75, 2011.

[3] V. Henzl, M. Swinhoe, S. Tobin and H. Menlove, "Measurement of the Multiplication of a Spent Fuel Assembly with the Differential Die-Away Method Within the Scope of the Next Generation Safeguards Initiative Spent Fuel Project," *Journal of Nuclear Materials Management*, vol. XL, no. 3, pp. 61-69, 2012.

[4] V. Henzl, M. Swinhoe, S. Tobin, H. Menlove, J. Galloway and D. Won Lee, "Direct Measurement of Initial Enrichment, Burn-up and Cooling Time of Spent Fuel Assembly with a Differential Die-Away Technique Based Instrument," in *Institute of Nuclear Materials Management*, Orlando, 2012.

[5] P. Blanc, H. Menlove, S. Tobin, S. Croft and A. Favalli, "An Integrated Delayed-Neutron, Differential-Die Away Instrument to Quantify Plutonium in Spent Nuclear Fuel," *Journal of Nuclear Materials Management*, vol. 40, no. 3, pp. 70-77, 2012.

[6] D. Pelowitz, "MCNPX User's Manual, Version 2.7.0," Los Alamos National Laboratory, LA-UR-11-00438, 2011.

[7] J. Galloway, H. Trellue, M. Fensin and B. Broadhead, "Design and Description of the NGSI Spent Fuel Library with an Emphasis on Passive Gamma Signal," *Journal of Nuclear Materials Management*, vol. 40, no. 3, pp. 25-35, 2011.

[8] T. Lee, H. Menlove, M. Swinhoe and S. Tobin, "Monte Carlo Simulations of differential die-away instrument for determination of fissile content in spent fuel assemblies," *Nuclear Instruments and Methods in Physics Research A*, vol. 652, pp. 103-107, 2011.

[9] T. Martinik, V. Henzl, S. Grape, S. Jacobsson Svard, P. Jansson, M. Swinhoe and S. Tobin, "Simulation of Differential Die-Away instrument's response to asymmetrically burned spent nuclear fuel," *submitted for publication in Nuclear Instruments and Methods in Physics Resarch A*, 2014.